

Effect of higher operating pressure on the net change in voltage of a proton exchange membrane fuel cell under various operating conditions

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Abstract

The performance improvement of a proton exchange membrane fuel cell due to increased operating pressure must be balanced against the energy required to pressurize the reactant gases. This paper quantifies and assesses the effect of higher operating pressure on the net change in cell voltage at various operating temperatures, air stoichiometries, relative humidities, and humidity ratios. The analysis is performed to determine the power lost due to air compression at various operating current densities. The calculated results at high-pressure ratios demonstrate the significance of a higher operating temperature and a lower air stoichiometry on the net change in the cell voltage. Furthermore, better results are achieved at a humidity ratio of 0.10 than at 0.05 in terms of net change in the cell voltage, relative humidity, and stoichiometric ratio.

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1. Introduction

From a thermodynamics viewpoint, the maximum work achieved in an electrochemical process is equal to the change in the Gibbs energy as the reactant converts product [1]. Based on the ideal gas law, the change in the Gibbs energy, ΔG , is related to the number of moles, gas constant, temperature and pressure. At a constant operating temperature and at different pressure, ΔG would be expressed in terms of the natural log of the pressure ratio. Thus, the higher the pressure ratio, the greater ΔG would be in the electrochemical reaction.

Technically, the performance of a proton exchange membrane (PEMFC) fuel cell in terms of voltage and power density can be greatly influenced by the operating pressure. Key parameters that contribute to the performance, such as temperature and air stoichiometry, can also be strongly affected by the operating pressure [2]. Moreover, enhancement of the electro-osmotic drag between the cathode and anode in the

fuel cell through a higher operating pressure difference across the membrane will result in better performance curves [3,4]. The performance can be adversely affected at extreme conditions such as high operating pressure, stoichiometric ratio, or cell temperature. Therefore, the improvement of the performance of the fuel cell due to increased pressure must be balanced against the energy required to pressurize or compress the reactant gases.

Humidity ratio, which represents the mass of water that exists in the air to fulfil the required humidity, does not give a clear picture of the drying effect in the fuel cell. Nevertheless, it plays a key role in the pressurization process of the reactant air since the required amount of water in air will be less at high pressure in order to achieve the same humidity. Thus, a very low humidity ratio could pose a major obstacle in having a high cell voltage. Unlike the humidity ratio, the relative humidity, which is defined as the ratio of the partial pressure of water to the saturated vapour pressure of water, is used to describe the drying effect of air in the fuel cell. For example, relative humidity at extreme conditions (i.e., very low or very high relative humidity) can lead to a lower

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Nomenclature

A	active fuel-cell area (cm^2)
c_p	specific heat of air ($\text{kJ kg}^{-1} \text{ }^\circ\text{C}^{-1}$)
F	Faraday's constant (96487 C eq.^{-1})
ΔG	change in Gibbs energy (kJ)
i	average current density (A cm^{-2})
I	current (A)
\dot{m}	mass flow rate (kg s^{-1})
\dot{m}_{air}	mass flow rate of inlet air (kg s^{-1})
M	molar mass (kmol kg^{-1})
n	number of cells
P	pressure (atm)
P_{FC}	fuel-cell power output (W)
P_{lost}	power lost (W)
\dot{P}_{lost}	power lost per surface area per cell ($\text{W cm}^2 \text{ cell}$)
R	gas constant ($82.06 \text{ cm}^3 \text{ atm mol}^{-1} \text{ K}^{-1}$)
T	cell operating temperature ($^\circ\text{C}$)
V_{FC}	fuel-cell voltage (V)
ΔV	net change in voltage (V)
X_{O_2}	mole fraction of oxygen in air

Greek symbols

γ	specific heat ratio
η	efficiency
λ	air stoichiometry
ϕ	relative humidity
ω	humidity ratio

Subscripts

a	air
c	compressor
gain	gain
loss	loss
m	electric drive (motor)
sat	saturated
t	total
w	water

fuel-cell performance as a result of a respective membrane dryness or membrane and electrode flooding. Therefore, it is extremely essential to quantify the influence of a high operating pressure on the fuel-cell performance at various operating temperatures, stoichiometric ratios, humidity ratios and relative humidities of air by taking into account the net voltage gain and voltage loss due to the pressurization process.

To date, numerous studies of the effect of a higher operating temperature, pressure and stoichiometric ratio on fuel-cell performance have been reported [3,5–7]. For instance, Springer et al. [5] carried out an experimental and theoretical study on a fuel cell at various relative humidities and operating temperatures, while Wakizoe et al. [6] and Kim et al. [7]

performed a similar analysis at variable operating conditions. On the other hand, Nguyen [8], Kazim et al. [9] and Wang and Liu [10] conducted theoretical and experimental studies on the effect of a higher operating temperature, pressure and stoichiometric ratio on a different type of flow field, namely, an interdigitated flow field. Others carried out exergy analysis on a PEMFC at different operating conditions [11]. In addition, De Francesco and Arato [12] investigated the transient response of a PEMFC and a compressor system used in automotive applications and under stable conditions with air relative humidity and temperature maintained in a proper range of values.

With the exception to the work of Natarajan and Nguyen [13], it should be pointed out that none of the above studies explicitly addressed the physical quantification of the voltage gain, or of the voltage losses that are associated with compressing the reactant air in the fuel cell. Therefore, the objective of the present work is to conduct a technical quantification and assessment of the effect of a higher operating pressure on the net change in the fuel-cell voltage at various operating temperatures, air stoichiometries, humidity ratios, and relative humidities. Moreover, the analysis is to be performed to determine the power lost due to compression at various current densities.

2. Model description

Generally, a higher operating pressure in a PEMFC will result in higher performance curves for voltage and power density. The voltage gain associated with increasing the operating pressure from an original value of P_1 to a higher pressure P_2 , is expressed as [14]

$$\Delta V_{\text{gain}} = \frac{RT}{4F} \ln \left(\frac{P_2}{P_1} \right) \quad (1)$$

The value of $RT/4F$ represents the influence of pressure on the exchange current density i_0 and is expressed in terms of the gas constant R , temperature T and Faraday's constant F . For simplicity, Hirschenhofer et al. [15] used a constant value of 0.06 V. In this work, however, exact calculations of the above term will be performed since it could deviate markedly from 0.06 V at high-pressure ratios, especially at $P_2/P_1 \geq 5$.

During the compression process, power will be lost as a result of pressurizing the reactant air to a higher pressure. The power lost is expressed in a form of electric drive efficiency η_m , compressor efficiency η_c , mass flow rate of air \dot{m}_{air} , specific heat ratio γ , and air specific heat c_p [14], i.e.,

$$P_{\text{lost}} = c_p \left(\frac{T}{\eta_m \eta_c} \right) \left(\left(\frac{P_2}{P_1} \right)^{\gamma-1/\gamma} - 1 \right) \dot{m}_{\text{air}} \quad (2)$$

The mass flow rate of the air utilized in the fuel cell during operation is related to the fuel-cell voltage V_{FC} and the power

output P_{FC} , as follows:

$$\dot{m}_{air} = \frac{M_a \lambda P_{FC}}{X_{O_2} F V_{FC}} \quad (3)$$

where M_a , λ and X_{O_2} are molar mass of air, air stoichiometry and mole fraction of oxygen in air, respectively.

Simply, the fuel-cell power is a product of operating current density i , fuel-cell voltage V_{FC} , active cell area A , and number of cells in the stack n , i.e.,

$$P_{FC} = i A n V_{FC} \quad (4)$$

Substituting Eqs. (3) and (4) into (2) at $M_a = 28.97 \times 10^{-3} \text{ kg mol}^{-1}$ and $X_{O_2} = 0.21$, and with values of c_p and γ are taken from Cengel and Boles [16] to yield the power lost in a more explicit form

$$\begin{aligned} P_{lost} &= 3.58 \times 10^{-4} \left(\frac{T}{\eta_m \eta_c} \right) \left(\left(\frac{P_2}{P_1} \right)^{0.286} - 1 \right) \\ &\quad \times \left(\frac{\lambda (i A n V_{FC})}{V_{FC}} \right) \\ &= 3.58 \times 10^{-4} \left(\frac{T}{\eta_m \eta_c} \right) \left(\left(\frac{P_2}{P_1} \right)^{0.286} - 1 \right) (\lambda i A n) \end{aligned} \quad (5)$$

The power lost per surface area per cell \dot{P}_{lost} can be expressed as

$$\begin{aligned} \dot{P}_{lost} &= \frac{P_{lost}}{n A} \\ &= 3.58 \times 10^{-4} \left(\frac{T}{\eta_m \eta_c} \right) \left(\left(\frac{P_2}{P_1} \right)^{0.286} - 1 \right) (\lambda i) \end{aligned} \quad (6)$$

Simply, the power lost described in Eq. (5) is the product of a voltage loss due to driving the compressor ΔV_{loss} and the operating current such that [14]:

$$\begin{aligned} P_{lost} &= I \Delta V_{loss} \\ &= [i A n] \underbrace{\left[3.58 \times 10^{-4} \left(\frac{T}{\eta_m \eta_c} \right) \left(\left(\frac{P_2}{P_1} \right)^{0.286} - 1 \right) \lambda \right]}_{\Delta V_{loss}} \end{aligned} \quad (7)$$

Clearly, the net change in cell voltage ΔV , which is the difference between the voltage gain and the voltage loss, can indicate whether the pressure increase will improve or kinder the performances of the fuel cell:

$$\Delta V = \Delta V_{gain} - \Delta V_{loss} \quad (8)$$

Depending on the magnitudes of ΔV_{gain} and ΔV_{loss} , the value of ΔV could take a positive or negative magnitude as the operating pressure is raised from P_1 to P_2 , as illustrated in Fig. 1. Therefore, the adverse effect of the pressurization process must also be taken into consideration when attempting to improve the performance of the fuel cell by increasing the operating pressure.

Generally, air humidification can add a great deal of importance to the pressurization (compression) of the reactant air in the fuel cell. The humidity ratio, which is the ratio of the rate of water production over the rate of exit airflow, can be expressed as [14]:

$$\omega = \frac{\dot{m}_w}{\dot{m}_a} = \frac{9.34 \times 10^{-8}}{3.57 \times 10^{-7} \times \lambda - 8.29 \times 10^{-8}} \quad (9)$$

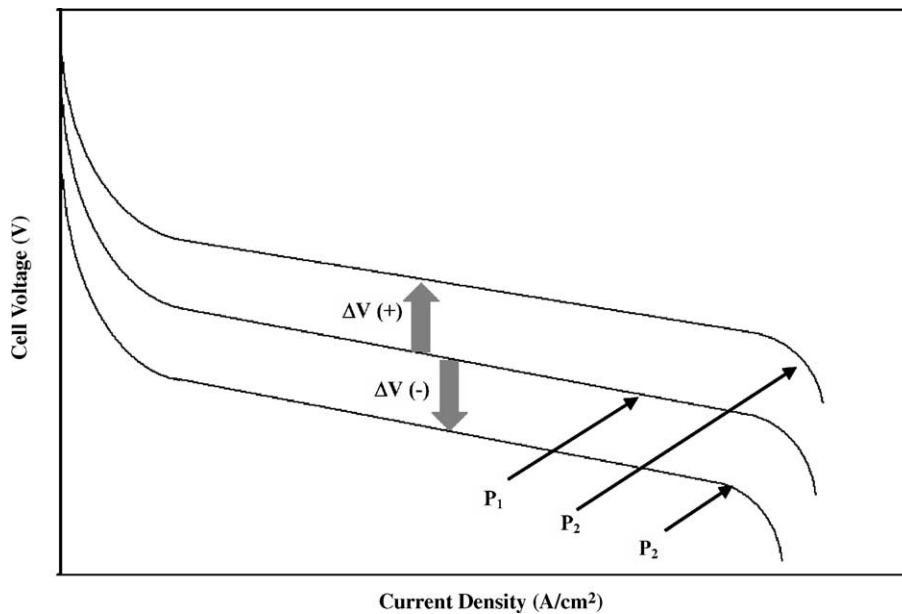


Fig. 1. Net change in fuel-cell voltage between P_1 and P_2 .

Similarly, the relative humidity ϕ , which is the ratio of the partial pressure of water over the saturated vapour pressure of water, can be described as

$$\phi = \frac{P_w}{P_{\text{sat}}} \quad (10)$$

At a fully humidified condition ($\phi = 100\%$), the partial pressure of water becomes equal to the saturated vapour pressure at a given operating temperature, i.e.,

$$P_w = P_{\text{sat}@T} \quad (11)$$

The present analysis will be conducted at two operating temperatures, namely, 60 and 80 °C. The respective partial pressures of water, P_w at a fully humidified condition are evaluated to be 19.94 and 47.39 kPa [16].

The total pressure of air P_t , is the sum of the partial pressure of dry air P_a , and the partial pressure of saturated water vapour P_w [16], i.e.,

$$P_t = \left(\frac{P_2}{P_1} \right) P_1 = P_a + P_w \quad (12)$$

Assuming the product water to evaporate completely and the water vapour and air to behave as an ideal gas, the humidity ratio can be re-described in the following form [16]:

$$\omega = \frac{M_w P_w}{M_a P_a} = 0.622 \frac{P_w}{P_a} = 0.622 \frac{P_w}{P_t - P_w} \quad (13)$$

Eq. (13) evaluates the effect of the fuel-cell pressure ratio on the humidity ratio, which is used to determine the air stoichiometric ratio from Eq. (9). Henceforth, the voltage loss is calculated from Eq. (7) at the specified operating conditions.

Through combining Eqs. (10), (12) and (13), an explicit relation between the relative humidity and the humidity ratio, can be established, namely:

$$\phi = \frac{\omega(P_2/P_1)P_1}{(0.622 + \omega)P_{\text{sat}}} \quad (14)$$

Table 1

Properties of fuel-cell system under standard conditions [14,16]

Property	Value
Original operating pressure, P_1 (atm)	1
Specific heat of air, c_p (kJ kg ⁻¹ °C ⁻¹)	1.005
Electric drive efficiency, η_m (%)	90
Compressor efficiency, η_c (%)	70
Specific heat ratio of air, γ	1.4
Molar mass of air, M_a ($\times 10^{-3}$ kg mol ⁻¹)	28.97
Molar mass of water, M_w ($\times 10^{-3}$ kg mol ⁻¹)	18.016
Gas constant, R (cm ³ atm mol ⁻¹ K ⁻¹)	82.06
Mole fraction of oxygen in air, X_{O_2}	0.21

It should be stressed that this work will be performed at a pressure ratio that range from $P_2/P_1 = 1$ to $P_2/P_1 = 20$. Furthermore, the analysis will be conducted at selected relative humidities of $\phi = 70$ and 100% (fully humidified condition), and at selected humidity ratios of $\omega = 0.05$ and 0.10. The properties of the fuel-cell system, as well as the standard operating conditions are presented in Table 1.

3. Results and discussion

This work describes the effect of a higher pressure ratio on a PEMFC at various operating conditions, in the form of net change in the fuel-cell voltage ΔV . The data presented in Fig. 2 show that a higher net change in the voltage at a lower cell temperature is observed due to smaller losses associated with air compression, which result in a smaller ΔV_{loss} as given by Eq. (7). Furthermore, ΔV increases with higher pressure ratios and at any cell temperature since the magnitude of the net voltage gain ΔV_{gain} would be greater than the net voltage loss ΔV_{loss} . For instance, a 0.1 V increase in the net cell voltage is observed at 60 °C as the operating pressure ratio is raised from $P_2/P_1 = 1$ to 10. It should be noted, however, that the difference in ΔV between various cell temperatures is

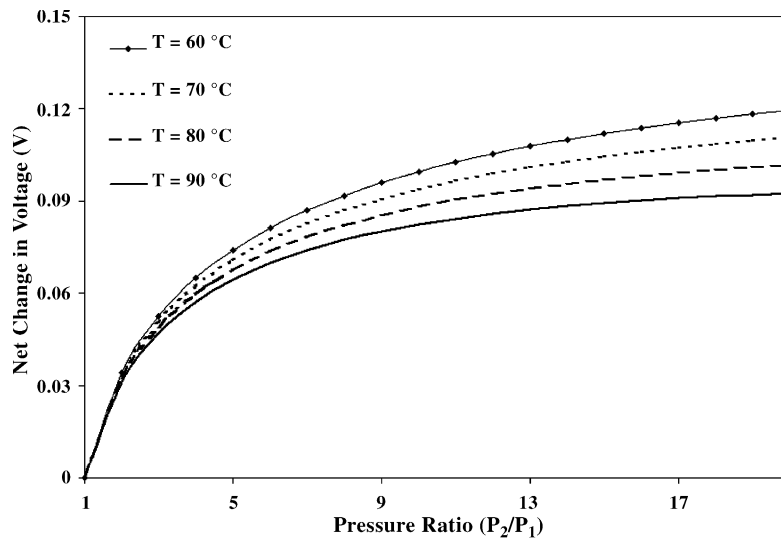


Fig. 2. Net voltage change vs. pressure ratio at various operating cell temperatures and at $\lambda = 2$.

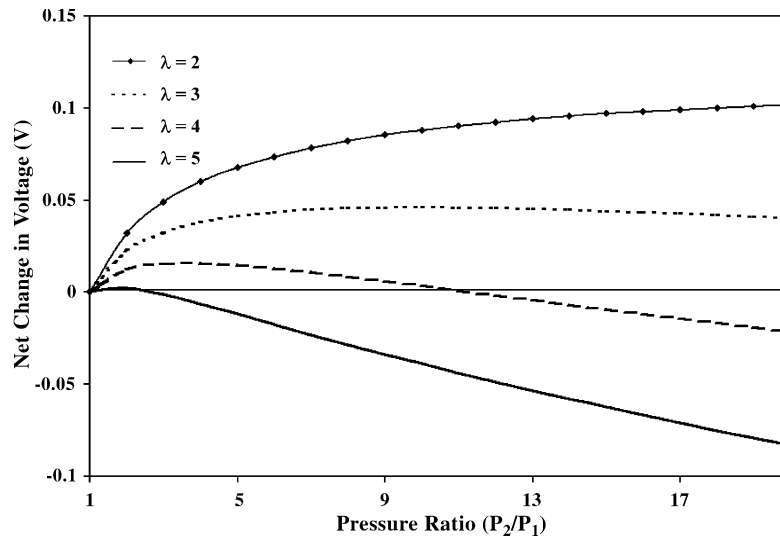


Fig. 3. Net voltage change vs. pressure ratio at various stoichiometric ratios and $T = 80^\circ\text{C}$.

considered to be more prominent than at low-pressure ratios (i.e., $P_2/P_1 \leq 3$). Thus, it is recommended to operate the fuel cell at a higher pressure to achieve a greater net change in the cell voltage and hence obtain better performance curves.

Depending on the air stoichiometry λ , the performance of the fuel cell can be greatly improved since the reactant mass flow rate of air is a strong function of its stoichiometry, described in Eq. (3). The net change in cell voltage at a low air stoichiometry (i.e., $\lambda \leq 3$) increases on increasing the pressure ratio, as depicted in Fig. 3. Conversely, at a high air stoichiometry ($\lambda \geq 4$), the net change in the cell voltage would take a negative value as the pressure ratio is raised from 1 to 20.

Generally, improvement in fuel-cell performance can be accomplished if the air stoichiometry in the fuel cell is raised

from 2 to 4. Nevertheless, caution should be exercised when setting up the air stoichiometry to be different from that of the recommended range of 2–4. This is mainly because at a high operating temperature and air stoichiometry ($\lambda > 4$), the relative humidity at the exit air will be lowered and thereby give rise to a higher risk for the cells to dry-out and a sharp decrease in fuel-cell efficiency would occur [11]. For example at $\lambda = 5$, a higher magnitude of ΔV_{loss} would take place compared with ΔV_{gain} as the pressure ratio is increased from 1 to 20, and this would result in a significant drop in ΔV .

At any given current density, the power lost per surface area per cell, \dot{P}_{lost} , varies exponentially with the fuel-cell pressure ratio, as shown in Fig. 4. In addition, greater \dot{P}_{lost} is expected at a higher operating current density, which varies linearly with the power lost in the fuel cell. For instance, the

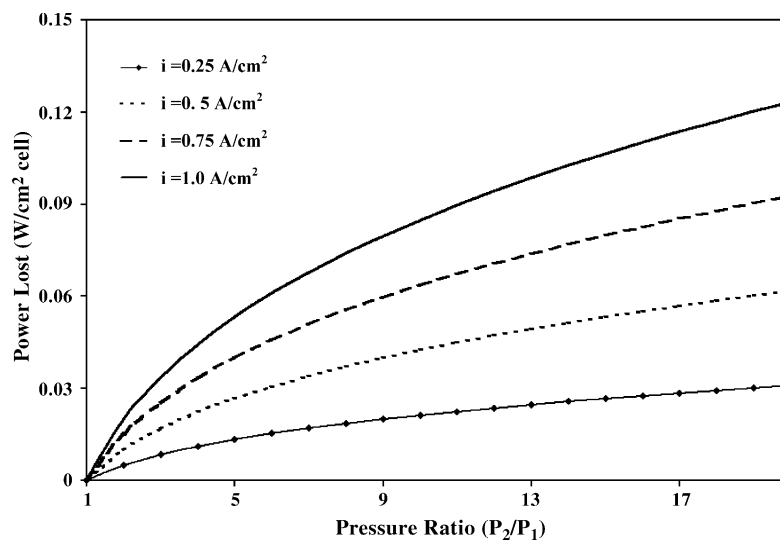


Fig. 4. Lost power vs. pressure ratio at various current densities and at $T = 80^\circ\text{C}$ and $\lambda = 2$.

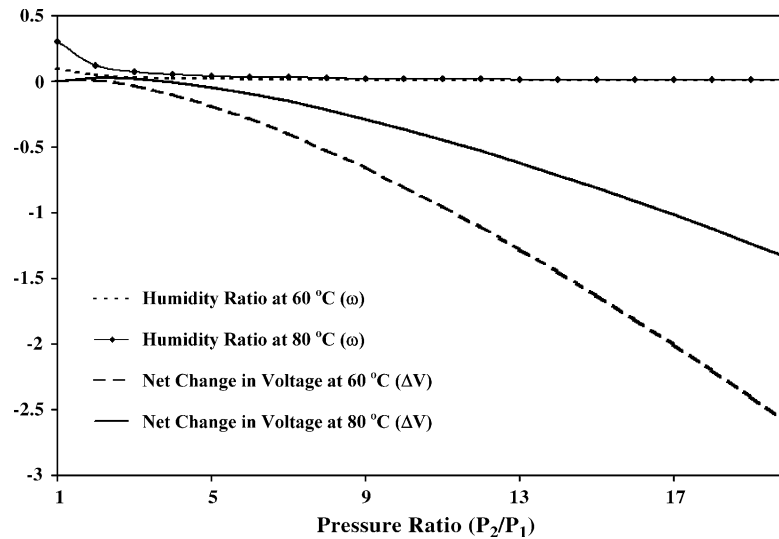


Fig. 5. Net voltage change and humidity ratio vs. pressure ratio at 60 and 80 °C and at $\phi = 70\%$.

\dot{P}_{lost} at a current density of $i = 1.0 \text{ A cm}^{-2}$ is four times greater than at $i = 0.25 \text{ A cm}^{-2}$. Similar conclusions can be drawn at different operating temperatures or stoichiometric ratios of air, as well as at any specific current density described in Eq. (6).

At low-pressure ratios, the difference in the humidity ratio between 60 and 80 °C tends to be far greater than at higher pressure ratios especially at $P_2/P_1 \geq 7$, as depicted in Fig. 5. This is mainly attributed to the value of the partial pressure of air, which represents the difference in magnitude between the total pressure and the pressure of the saturated water. For example, the partial pressure of air at 80 °C and at $P_2/P_1 = 20$ is 23 times greater than at $P_2/P_1 = 1$ causing a substantial reduction in ω from 0.30 to 0.01 at $P_2/P_1 = 20$. By the same token, a major drop in the net change in the cell voltage at a lower temperature, i.e., at 60 °C is detected as a result of

a lower air mass flow rate and hence a higher stoichiometric ratio would be required to complete the reaction and would result in a larger ΔV_{loss} .

Similar remarks can be made for a fuel cell operating at a fully humidified condition ($\phi = 100\%$) as presented in Fig. 6. In fact, better results are achieved at this condition than at $\phi = 70\%$, in terms of net change in the cell voltage, stoichiometric ratio and humidity ratio. For instance, the humidity ratio of a fully humidified cell operating at $P_2/P_1 = 5$ and at 60 °C is 30% higher than at $\phi = 70\%$. In addition, the ΔV of a fully humidified fuel cell operating at $P_2/P_1 = 5$ and a temperature of 80 °C is 0.0063 V, which is about seven times greater than at $\phi = 70\%$. The reason behind this increase is that the calculated stoichiometric ratio at $\phi = 70\%$ is $\lambda = 6.24$, which is approximately 45% higher than at $\phi = 100\%$ and hence causes a higher ΔV_{loss} .

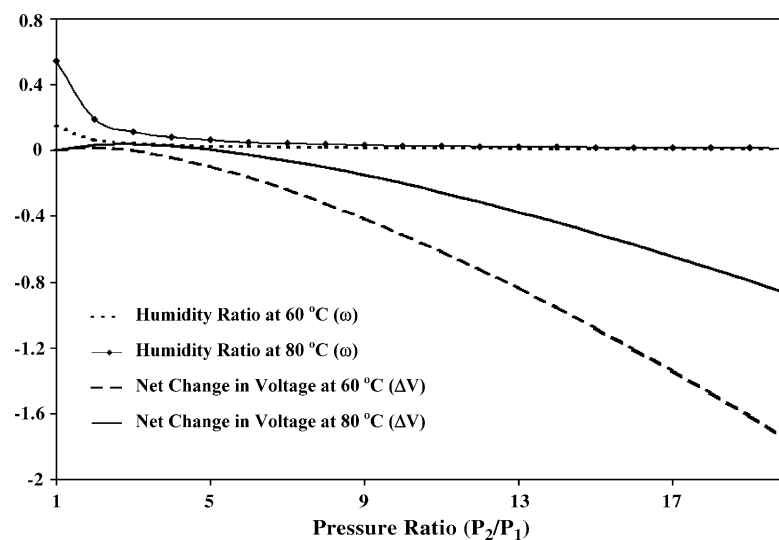


Fig. 6. Net voltage change and humidity ratio vs. pressure ratio at 60 and 80 °C and at a fully humidified condition of $\phi = 100\%$.

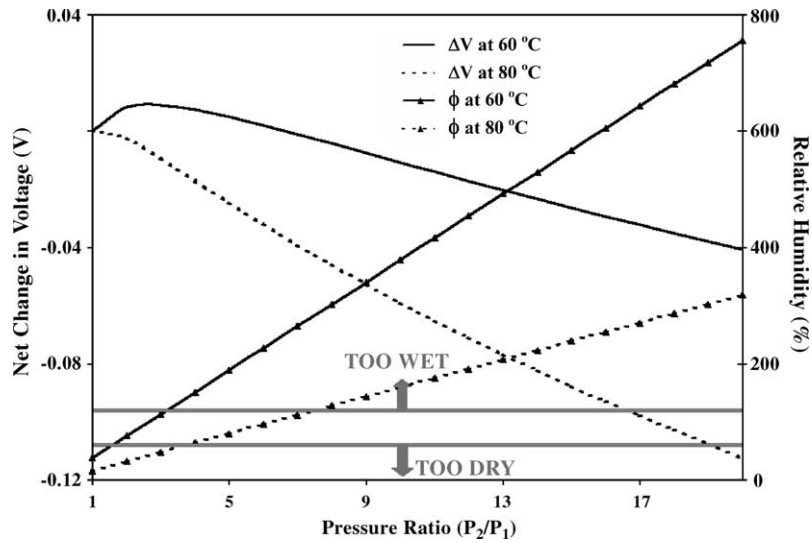


Fig. 7. Net voltage change and relative humidity vs. pressure ratio at $\omega = 0.05$.

There is usually a strong relation between the relative humidity of a fuel cell and its performance, such as the net change in the cell voltage shown in Fig. 7. At a given humidity ratio of $\omega = 0.05$, the relative humidity at 80°C is 60% lower than at 60°C . Hence, a lower net change in cell voltage would take place. Moreover, it is observed that ΔV at 60°C takes a positive magnitude as P_2/P_1 varies from 1 to 6, then drops to a negative value at a pressure ratio beyond that range. On the other hand, ΔV at 80°C displays a continuous negative trend as P_2/P_1 varies from 1 to 20, where $\Delta V = -0.11\text{ V}$. From these results, it is calculated that it would be highly essential to have extra humidification for a fuel cell operating above 60°C [17].

It should be emphasized that a relative humidity greater than 100% is impossible to achieve in reality. Moreover, the fuel-cell electrodes will be completely flooded if the theoretical relative humidity is much greater than 100%. Conversely, if the fuel cell operates at a low relative humidity (i.e., $\phi \leq 70\%$), an adverse effect on its membrane and electrode assemblies could occur as a result of a complete cell dry-out and hence a lower voltage and power density would be anticipated. Therefore, it is important to maintain cell humidification in the recommended theoretical range of $80 \leq \phi \leq 120\%$ [14].

Contrary to the case at which $\omega = 0.05$, the net change in the voltage of a fuel cell operating at 60 and 80°C would take positive magnitudes at a humidity ratio of $\omega = 0.10$, as

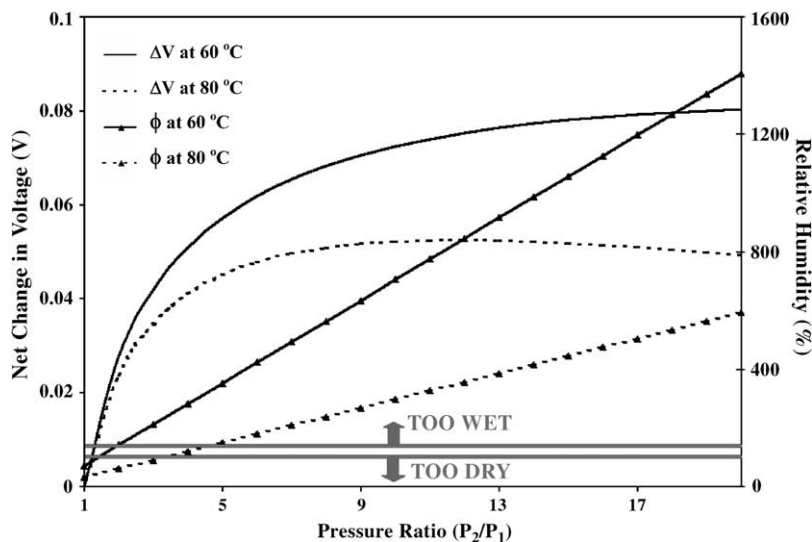


Fig. 8. Net voltage change and relative humidity vs. pressure ratio at $\omega = 0.10$.

depicted in Fig. 8. This is because the calculated stoichiometric ratio at $\omega = 0.10$ is $\lambda = 2.85$, which is half of the stoichiometric ratio at $\omega = 0.05$, and results in a lower ΔV_{loss} . In addition, an insignificant difference in ΔV between the two cell operating temperatures is observed at $P_2/P_1 \leq 5$ since the differences in ΔV_{gain} as well as ΔV_{loss} between the two temperatures at this low-pressure ratio are almost negligible, and therefore result in a negligible ΔV , as explained earlier. Similar to the previous case, linear trends of relative humidity versus pressure ratio at both operating temperatures of 60 and 80 °C are illustrated. Also, it should be noted that the calculated relative humidity at $\omega = 0.10$ and at any selected pressure ratio is approximately twice that at $\omega = 0.05$, since the relative humidity in Eq. (14) is strongly influenced by ω in the nominator than in the denominator.

Typically, the operating pressure of a fuel cell is reported to vary from 1 to 5 atm [2,3,9,10,18]. The present study, however, covered a wider range of a cell pressure ratio specifically from 1 to 20, which is considered to be far greater than any experimental or theoretical analysis performed to date on a PEMFC. The reason behind selecting this range was to quantify and assess the fuel-cell performance under extreme operating conditions (i.e., extremely high pressure) and to determine its parameters associated with such conditions. Moreover, the work predicts that the net change in cell voltage could be improved by at most 15% if the electric drive and compressor efficiencies are increased by 5%, although the values used here are considered to be realistic and reflect the actual efficiencies during the pressurization process.

4. Conclusions

In the present study, the effect of a higher operating pressure on the net change in cell voltage at various operating temperatures, air stoichiometries, humidity ratios and relative humidities has been quantified and assessed. The conclusions are as follows:

- A higher operating temperature could contribute significantly to the net change of the fuel-cell voltage at high-pressure ratios.
- Lower air stoichiometry would result in a greater net change in cell voltage. It is recommended, however, that the fuel cell be operated with an air stoichiometry range

between 2 and 4 in order to avoid any cell dry-out, which would decrease performance.

- Power lost per surface area per cell during the compression process, varies exponentially with the fuel-cell pressure ratio at any given current density.
- Contrary to the net change in cell voltage, the difference in the humidity ratio at low-pressure ratios tends to be far greater than at higher pressure ratios, especially at $P_2/P_1 \geq 10$.
- Better results are achieved at a humidity ratio of $\omega = 0.10$ than at $\omega = 0.05$ in terms of the net changes in cell voltage, relative humidity and stoichiometric ratio.
- The theoretical relative humidity of the cell should be in the recommended range of $80\% \leq \phi \leq 120\%$ in order to avoid either cell dry-out or flooding in the membrane and electrode assemblies.

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